

REMARKS

The above amendments and the following remarks are fully and completely responsive to the Office Action dated August 24, 2004.

Claims 1-25 are pending. Claims 1, 2, 4-9 and 12-19 are rejected. Claims 3, 10 and 11 are objected to. Claims 6-8, 13, 17-21 are amended. No new matter is added. All the claims are supported by at least the specification.

Specification

The specification is objected to for informalities. Applicants respectfully submit that the objection is obviated in view of the amendments indicated herein. Applicants urge withdrawal of the objections.

Allowable Subject Matter

Applicants respectfully acknowledge that claims 3, 10, 11, 20-24 are allowable over the prior art, subject to the objections to claims 3, 10, and 11, which are addressed below.

Claim Objections

Claims 3, 10 and 11 are objected to. Applicants respectfully acknowledge the objection and submit that the objection is obviated in view of the claim amendments indicated herein, in particular amendments to claim 1. Applicants request reconsideration and withdrawal of all objections.

Claim Rejections - 35 U.S.C. §101

Claims 17-19 are rejected under 35 USC §101 because the claimed invention is directed to non-statutory subject matter.

Applicants have rewritten claims 17-19 as product claims, thereby obviating the rejection under §101. Accordingly, Applicants request reconsideration and withdrawal of the rejection under 35 U.S.C. § 101 of claims 17-19.

Rejection under 35 U.S.C. § 103(a)

Claims 1, 2, 4-9 and 12-16 are rejected under 35 U.S.C. § 103(a) as being unpatentable over U.S. Patent No. 6,146,747 to Wang et al. ("Wang") in combination with U.S. Patent No. 5,091,086 to Stengaard ("Stengaard"), U.S. Patent No. 6,179,132 to Moya ("Moya") and WO 00/61267 to Kidd ("Kidd").

Applicants traverse this rejection. Applicants note that the technical problem addressed by the present invention, as discussed, for example, on pages 5 and 6 of the original specification, is to have available hydrophilic membranes for fuel cells showing the following combination of properties:

- improved water permeability;
- easily wettable;
- controlled porosity to gases;
- high conductivity in the cells; and
- operable also with hydrogen from reforming.

The Applicants have found porous hydrophilic membranes, as defined in the claims, which solve the above-mentioned technical problem. For example, the membranes of the invention, as defined in claim 1, comprise a porous support on which an amorphous ionomer in acid form is deposited.

Applicants further describe exemplary embodiments of the presently claimed invention through the examples presented in the specification.

Example 2 (on page 29 of the specification) discloses the preparation of a crosslinked membrane containing 26% of an ionomer, according to the invention, for use in fuel cells. The ionomer equivalent weight (EW) is 461. It is found that water permeability ($\text{liter}/(\text{h} \cdot \text{m}^2 \cdot \text{Atm})$) of said membrane is 25.

Example 3 (on page 31 of the specification) discloses the performance of the membrane of Example 2 in a fuel cell. The current density (A/cm^2) has been found to be 0.37 at 0.7 V and 0.85 at 0.6 V.

Comparative Example 5 (on page 33 of the specification) discloses the preparation of a membrane containing 22% by weight of an ionomer having a crystallinity of 11% (page 32, last line). Thus, the comparative membrane has an ionomer content comparable to that of example 2.

Applicants note that the membrane prepared in Example 2 (on page 29) is transparent in the dry state, and therefore, it is wettable. Both membranes were occluded to gases (see page 31, line 1 and page 33, line 18). Applicants note that the membrane of comparative Example 5 was found to be also impermeable to water (page 33, line 19 of the original specification).

Comparative Example 6 illustrates the performance in a fuel cell of the membrane of comparative Example 5, wherein at 0.7 V, the current density was of 0.21 A/cm² and at 0.6 V of 0.52 A/cm². Applicants note that although the ionomer content of the membrane, as shown in comparative Example 5, is comparable to that of the membrane of Example 2 (22% as compared to 26%), when operated in a fuel cell, the membrane of the comparative examples yields a current density about 40% lower at both 0.7 V and at 0.6 V.

Example 9 (on page 37 of the original specification) shows that a membrane containing an amorphous ionomer (from example 8 on page 35 of the original specification), which provided the same current density at 0.7 and 0.6 V as in the membrane from comparative Example 6, contains 13% weight of amorphous ionomer. Applicants note that the membrane as shown in Example 9, contains nearly one-half the ionomer content as that of the comparative membrane.

Applicants emphasize that in the present invention, wherein the produced current density is equal to that of the comparative membrane, the ionomer content of the membrane containing the amorphous ionomer is nearly half the ionomer content of the comparative membrane. Thus, there is a clear advantage of using a membrane, as presently claimed, of an amorphous ionomer in a fuel cell. Further, the membrane used in example 9, unlike that shown in the comparative example, shows water permeability. See also, Example 8, page 37, line 10.

Applicants submit that the presently claimed invention is distinguishable from each of the above-cited references. Applicants further submit that the references, either alone or in combination, do not teach or suggest the presently claimed invention.

Applicants submit that the presently claimed invention is clearly distinguishable from Wang. Wang discloses in the abstract synthetic polymeric materials formed from casting PVDF polymer solutions and/or dispersions. Said membranes are highly porous and are useful in a variety of microfiltration and ultrathin applications. Wang discloses that a membrane that is hydrophobic after casting may be treated with a wetting agent to render it hydrophilic. See Wang, column 14, lines 53-55. Wang further discloses that internally hydrophilic membranes may contain about 0.5% to about 4% of high molecular weight PVP. See Wang, column 16, lines 15-17.

Applicants point out, however, that Wang is unrelated to the presently claimed invention. Wang does not disclose that the membrane contains an ionomer compound, as required in the present claims. Accordingly, the Office Action *incorrectly* asserts on page 3, lines 14-16 that "Wang et al. does not disclose the water permeability of higher than $1 \text{ l/m}^2 \cdot \text{atm} \cdot \text{cm}$ or that the ionomer is of amorphous form and having a hydrophilic group on the acid form." Applicants note that Wang is completely silent on membranes containing ionomers.

Applicants submit that Stengaard does not make up for the deficiencies of Wang. Stengaard discloses permeable, porous polymeric membranes with

hydrophilic character. The hydrophilic character is obtained "by treatment with a solution comprising one or more hydrophilic, mono- or polymeric compounds selected among soluble, OH-containing cellulose derivatives, polyvinyl alcohols and low molecular weight, polyfunctional, NH- and/or OH-containing compounds, optionally in the presence of a crosslinking agent, a surfactant and an initiator." See Stengaard, Abstract. The layer deposited on the surface of the membrane is rendered insoluble by means of a catalyst reaction at elevated temperatures in order to fixate the hydrophilic material to the membrane. The membranes to be treated are preferably PVDF-, CTFE/VF- or PVDF-CTFE/VF. See Stengaard, column 6, lines 27-29.

Applicants submit that Stengaard has the same deficiencies as Wang. In particular, Applicants point out that Stengaard is unrelated to the presently claimed invention. In particular, Stengaard does not disclose that the membrane contains an ionomer compound, as required in the present claims.

With respect to Moya, Applicants submit that Moya does not make up for the deficiencies of Wang and Stengaard. Moya discloses a porous membrane formed from a porous polyperfluorocarbon membrane substrate having its surface modified with a perfluorocarbon polymer composition, the modified surface is directly wet with an aqueous liquid. See Moya, Abstract. The perfluorocarbon polymer is generally a copolymer of at least two monomers, one monomer being at least selected from VF, HFP, VDF, trifluoroethylene, CTFE, PAVE, TFE, and the other containing hydrophilic functional groups that can be converted to (SO₂F), (SO₂M), SO₃R), (PO₃M), (COF), (CO₂M), (CO₂R) or

(CONR₂) groups, wherein M is H, an alkali metal, an alkaline earth metal or NR₄, and each R separately is H, an alkyl group or an aryl group, optionally substituted. See Moya, column 5, lines 47-62.

Moya also discloses that generally, sulfonyl, carbonyl, sulfonate and carboxylate esters and sulfonyl and carbonyl based amide forms of the perfluorocarbon polymers are readily converted to ion exchange forms by a hydrolysis reaction. See Moya, column 11, lines 14-17. Moya also discloses that the molecular weight of the perfluorocarbon polymer is between 250 and 1500. See Moya, column 11, lines 34-35. In example 1 (column 14) and example 3 (column 15), the perfluorocarbon polymer used to modify the membrane surface is Nafion®.

Applicants note that the technical problem addressed by Moya is to provide a porous membrane having its entire surface modified with a perfluorocarbon polymer including hydrophilic functional groups, so that the membrane surface is directly wettable with water. See Moya, column 5, lines 38-42. However, Moya does not address the technical problem addressed by the present invention, as Moya's main concern is to make membranes directly wet with an aqueous liquid.

Applicants also note that the Nafion® used in examples 1 and 3 is known to contain residual crystallinity. See the attached document, which discusses Nafion® Morphology. Based on this information, Applicants submit that Moya does not teach or suggest membranes containing amorphous ionomers, as in the presently claimed invention.

The Applicants have also shown that membranes coated with an ionomer showing crystallinity are impermeable to water and also have a lower performance in fuel cells. Thus, the results achieved by the present invention were surprising and unexpected, in light of the references such as Moya.

Applicants note that Kidd does not make up for the deficiencies of the previously discussed references. Kidd discloses integral membranes comprising an amorphous halopolymer. Moreover, Kidd discloses that the treatment of corrosive or reactive fluids can use composite membranes, wherein an amorphous fluoropolymer is disposed as a surface treatment or coating on the pore walls of a porous substrate. See Kidd, page 1, lines 22-23. Kidd also discloses that these membranes are not satisfactory for many applications.

Applicants note that the technical problem addressed by Kidd is to find a porous halopolymer membrane suitable for treating corrosive or reactive fluids. See Kidd, page 1, lines 28-29. As a result, Kidd discovered a solution involving an integral porous membrane comprising an amorphous polymer. See Kidd, page 2, lines 23-24. The halopolymers suitable for preparing said membranes have a degree of crystallinity less than about 30%. See Kidd, page 3, lines 11-13.

The monomers of the amorphous copolymer are fluorinated olefin monomers, for instance, TFE, hexafluoropropylene and chlorotrifluoroethylene, and fluorinated functional monomers, such as perfluoroalkylvinyl ethers, perfluoroesters, perfluorosulfonyl fluorides, perhalodioxoles and preferably perfluorodioxoles. See Kidd, page 3, lines 27-30 and page 4, line 1.

The preparation of the membranes comprises the following steps (page 6, lines 9-14):

- (a) providing a solution of the halopolymer;
- (b) shaping the solution to form a pre-membrane; and
- (c) causing phase inversion of the pre-membrane to obtain integral porous membranes.

The membranes of Kidd can be used as filters, particularly as gas filters, i.e., to allow gases to pass while retaining fluids. See Kidd, page 9, line 14 and page 10, lines 3-4.

Applicants note that by using the process disclosed by Kidd, membranes of amorphous polymers with ester or sulfonyl functional groups are obtained. Applicants further note that Kidd is completely silent on fuel cell applications of said membranes.

As previously noted, the technical problem addressed by Kidd is to find a porous halopolymer membrane suitable for treating corrosive or reactive fluids. Applicants submit that Kidd is not relevant to the present invention. Kidd is not concerned with the technical problem addressed by the present invention. Moreover, Kidd does not teach or suggest a solution, such as the solution recited in present claim 1, since the membranes of Kidd do not contain amorphous polymers in acid form.

Applicants refer to the Office Action (page 5, lines 13-16), which asserts that “the ionomer being amorphous and the hydrophilic group being in acid form, the prior arts of Stengaard, Moya and Kidd show that these aspects are known in

the art to be used in conjunction with the making of a porous hydrophilic membrane.”

In response, Applicants note that neither Stengaard nor Wang disclose any amorphous ionomer in acidic form. Furthermore, the technical problem addressed by the presently claimed invention is not limited to obtaining a membrane being porous hydrophilic, but to having a membrane showing the combination of properties related to the technical problem addressed by the present invention.

Accordingly, Applicants submit that Kidd fails to address the technical problem of the present invention and also fails to indicate the solution found by the Applicants. The same holds for Moya, which only discloses, as discussed above, membranes directly wettable with an aqueous liquid.

The Office Action (on page 5, line 16 to page 6, line 3) asserts that the membrane disclosed by Wang, to which a hydrophilic character could be imparted by treating the membrane according to Stengaard, would teach or suggest to one skilled in the art how to make a membrane hydrophilic in order to achieve a water permeability greater than $1 \text{ l/m}^2\text{h.atm}$.

Applicants disagree with such an assertion. Applicants again note that both Wang and Stengaard fail to suggest the technical problem of the present invention. Further, neither Wang nor Stengaard teaches or suggests membranes containing amorphous ionomers in acidic form.

Regarding dependent claims 2 and 4, the Office Action asserts that Wang shows that the ionomer is less than about 20% and that the membrane has a microporous surface with minimum pores. However, Applicants note that no amorphous ionomer in acid form is contained in the membrane of Wang, and that the other cited references do not make up for the deficiencies of Wang.

Although Wang (column 5, lines 59-64) discloses that said membranes can contain polyvinylpyrrolidone (PVP) in quantities up to 30% weight, Applicants note that PVP is not an ionomer compound.

Additionally, Applicants point out that the term "microporous," as used in Wang, relates to membrane surfaces with an average pore diameter exceeding about 0.01 μm and that membranes having an average pore diameter less than 0.01 μm are classified as gas separation membranes. See Wang, column 5, lines 1-3. The membranes of Wang are permeable to gases since they have an average pore diameter exceeding about 0.01 μm . Applicants therefore conclude that Wang is not relevant to present claims 2 and 4.

Applicants further note that dependent claims 5, 6-9 and 12-16 are patentable for the same reasons as claim 1. Thus, reconsideration and withdrawal of the rejection of claim 1, 2, 4-9, and 12-16 under 35 U.S.C. §103(a) are respectfully submitted.


Conclusion

In view of the amendments and remarks above, Applicants submit that this application is in condition for allowance and request favorable action thereon.

Should the Examiner believe anything further is desirable in order to place this application in better condition for allowance, the Examiner is requested to contact the undersigned at the telephone number listed below.

In the event this paper is not considered to be timely filed, Applicants respectfully petition for an appropriate extension of time. Any fees for such an extension, together with any additional fees that may be due with respect to this paper, may be charged to counsel's Deposit Account No. 01-2300, **referencing docket number 108910-00054.**

Respectfully submitted,


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Enclosure: One page document, discussing Nafion® Morphology


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**Morphology and
Morphological Changes
Within Nafion**

Membranes Induced by Mechanical Orientation

**Authors: Trevino, S. F.; Young, Sandra K.; ARMY
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Abstract: The structure of Nafion, a perfluorosulfonated ionomer, has been the subject of many studies. Morphology evaluations have used the methods of small-angle x-ray scattering (SAXS), wide-angle x-ray scattering (WAXS), and small-angle neutron scattering (SANS). Three principal features in the small-angle scattering pattern have been observed on the H⁺-form Nafion. A peak at approximately Q approx. 0.2 A⁻¹ is attributed to the clustering of the acid groups (ionomer peak), that at Q -0.03 A⁻¹ to crystalline regions, and an upturn in intensity at the smallest Q to large-scale heterogeneities. Some of the previous works have included samples that have experienced moderate strains by mechanical elongation. In those works, the effect on the ionomer peak has been studied. The WAXS studies led to a model of the packing of the polymer molecules into ionic aggregates, while the SAXS and SANS studies resulted in models of the packing of the ionic clusters. In the present work, SANS measurements on elongated samples of Nafion have been used to obtain data that suggests a model of the structure of the material. The samples studied were elongated at two temperatures, 25 deg. and 155 deg. C. Although many of the scattering features of these two samples are similar, the samples stretched at 155 deg. C contain new information concerning the nature of the crystalline region.

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